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A nonlinearity in the photoluminescence emission of single crystal CdS excited by He-Cd laser

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Abstract : Very intensive studies about photoluminescence spectra of semiconductor crystals have been done for a long time. When single crystal CdS is excited at low temperatures with photon energy above the band gap, electron-hole pairs are produced and these can recombine in a number of ways, some of which give rise to luminescence. Photoluminescence spectra of the single crystal CdS have been obtained at 5 and 7K using the low temperature photoluminescence experimental arrangement. He-Cd laser that has 325nm wavelength and 9mW output power was used as the excitation source and the experimental data obtained by computer-controlled spectrometer was processed by means of computer. Photoluminescence spectra in the range of 2.53 to 2.21 eV have been recorded and it has been seen from the experimental data as 5 and 7 K that there are some ratios between intensities of phonon replicas and intensities of donor-acceptor pair emission and this ratios give us nonlinear results in the spectral emission range of the CdS single crystal.

Keywords : Photoluminescence, cadmium sulfide crystal, He-Cd laser

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In recent years, there have been significant works about optical properties of both bulk and colloidal CdS at low temperatures. Due to their industrial applications, studies of the optical nonlinearity of CdS have gained importance. The works of nonlinearity recently have been focused on CdS single crystal [1] and the other materials including CdS [2] and also CdS doped silica films [3]. Moreover, properties of nonlinearity of CdS are getting more significant in optical switching [4]. Relating to the optical properties of CdS, photoluminescence studies of CdS have recently been concentrated on this films [5, 6], heterostructures [7], nanocrystals [8, 9] and also colloids [10]. The works of photoluminescence of electrochemically deposited CdS has been investigated at 77K and peak emission at 488 nm has been observed [11]. Phase transition and photoluminescence study of CdS thin film has been done. Well known additional green band [12] has been observed in CdS-$CdS_{1-x}Se_x$ superlattice [13]. In this work, photoluminescence spectra of CdS single crystal excited by He-Cd laser have been obtained. Analysing the emission spectra, the dominant peaks were identified and from the numerical ratios of the relative intensity of donor-acceptor peak to those of phonon replicas, a nonlinearity in the spectral range has been obtained by using Sigma Plot Scientific Graph System programme.

The experimental arrangement used in the photoluminescence experiment mainly consists of He-Cd laser, computer-controlled spectrometer, low temperature cryostat with liquid helium flow, photomultiplier tube, phase sensitive detector (lock-in amplifier), photodetector, optical chopper, concave lenses, mirror and computer including software and hardware related to photoluminescence measurement set-up. In the experimental study, first, the semiconductor sample was mounted in the cryostat so that front surface geometry would be at 45° to eliminate optical interference. Since the He-Cd laser emits photon of 325nm wavelength ($h\upsilon$ = 3.81 eV, $h\upsilon_{(He-Cd)} > E_{g(CdS)}$) and 9mW output power, the laser was used as an excitation source. Laser beam was focused by the concave lens onto the sample mounted in the cryostat. Optical filter was used to prevent the exciting laser light from entering the monochromator. Moreover, a mechanical chopper was used in order to modulate the light. Thus, photoluminescence measurements were done.

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Utilising the experimental set-up, the ambient temperature of the single crystal was decreased from room temperature to 5 K using liquid helium and photoluminescence emission in the range 2.21 to 2.53 eV was obtained as shown in Figure 1.

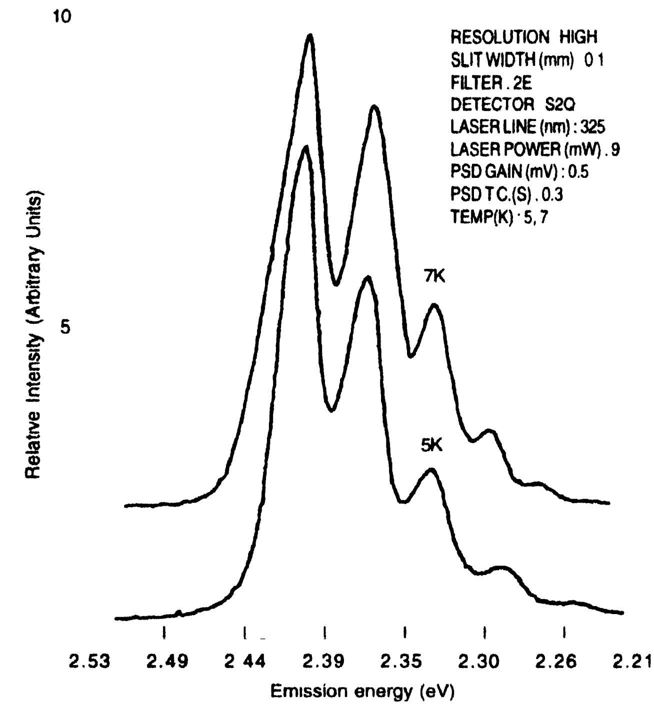

Figure 1. Photoluminescence spectra of CdS at 5 and 7 K.

As seen in Figure 1, the peaks at 2.40 eV correspond to donor-acceptor pair emission, the other four peaks (at 2.36, 2.32, 2.28 and 2.24 eV) to phonons [14-16] in the spectral emission. Experimental parameters used and the obtained data (in numerical values) are given in Table 1.

Using the numerical data given in Table 1, the ratios of the relative intensities of phonon replicas and that of donor-acceptor pair emission at 5 and 7 K were evaluated and third order nonlinear interpolation curves have been obtained as seen in Figure 2.

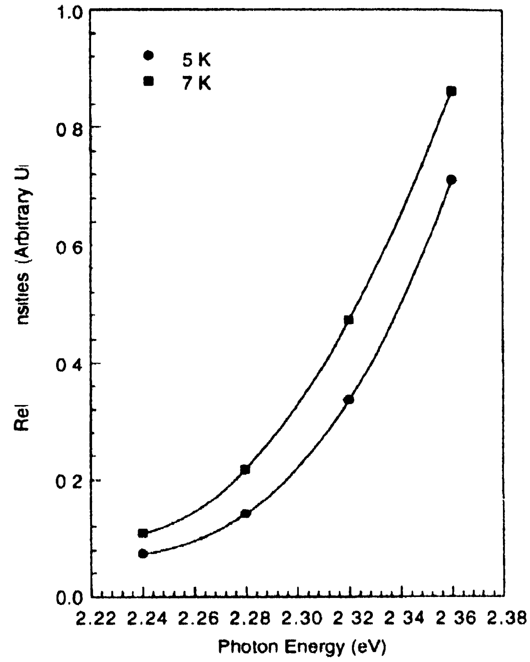

Figure 2. Plots of relative intensity ratios of the phonon replicas and the donor-acceptor band against photon emission energy at 5 and 7 K.

Concerning the numerical values of RRI (Ratio of Relative Intensities) and PhE (Photon Energy) of photoluminescence emission at 5 and 7 K, an empirical equation has been obtained as

$$RRI = b_0 + b_1.(PhE) + b_2.(PhE)^2 + b_3.(PhE)^3. \quad (1\ 1)$$

Table 1. Photoluminescence measurements of single crystal CdS

Experimental parameters	Temperature (K)	Phonon replicas energy (eV)	Peak of Donor-acceptor energy (eV)	Intensity of phonon replicas/donor-acceptor pair emission
Resolution : High		2.36		7.1/10
Slit width · 0.1 mm				
Filter : 2E (Wratten)		2 32		3.35/10
Detector : S2ϕ				
Laser line : 325 nm	5K	2.28	2.40	1 41/10
Laser output power : 9 mW				
PSD time constant · 0 3 s		2 24		0.74/10
PSD gain : 5mV				
Resolution : High		2.36		8.62/10
Slit width : 0.1 mm				
Filter : 2E (Wratten)		2.32		4.71/10
Detector : S2ϕ				
Laser Line : 325 nm	7K	2.28	2.40	2.17/10
Laser output power : 9 mW				
PSD time constant : 0.3 s		2.24		1.08/10
PSD gain : 5 mV				

Numerical details of the third order interpolation curves at 5 and 7 K with respect to eq. (1.1) are given below :

T = 5 K		T = 7 K	
Coefficients		Coefficients	
b[0]	1467.2100493814	b[0]	472.2720126952
b[1]	2015.1375644352	b[1]	5.2695418323e+2
b[2]	9.2218752802e+2	b[2]	187.8125072037
b[3]	140.6250040608	b[3]	-20.8333343773
PhE	**RRI**	**PhE**	**RRI**
2.24	0.0739999999	2.24	0.108
2.246	0.0780766251	2.246	0.11479175
2.252	0.0839780002	2.252	0.124999
2.258	0.0918863753	2.258	0.1385947499
2.264	0.1019840003	2.264	0.1555519999
2.27	0.1144531253	2.27	0.1758437499
2.276	0.1294760003	2.276	0.1994429999
2.282	0.1472348752	2.282	0.2263227499
2.288	0.1679120002	2.288	0.256456
2.294	0.1916896251	2.294	0.28981575
2.3	0.21875	2.3	0.326375
2.306	0.2492753749	2.306	0.36610675
2.312	0.2834479998	2.312	0.408984
2.318	0.3214501248	2.318	0.4549797501
2.324	0.3634639997	2.324	0.5040670001
2.33	0.4096718747	2.33	0.5562187501
2.336	0.4602559997	2.336	0.6114080001
2.342	0.5153986247	2.342	0.6696077501
2.348	0.5752819998	2.348	0.730791
2.354	0.6400883749	2.354	0.79493075
2.36	0.7100000001	2.36	0.862

These values given above show that there is a third order nonlinearity between the values of PhE and RRI of single crystal CdS at 5 and 7 K.

In this experimental work, photoluminescence measurement was carried out at liquid helium temperatures to prevent thermal ionisation of the optically active centres and to minimise the effect of lattice vibrations in the crystal. As in Figure 1, it has been seen that the peaks of phonon replicas are quite dominant in the spectra as well as donor-acceptor pair emission and also decreasing of intensity of the phonon replicas against the emission spectrum are very clear. Concerning the numerical data of the experiment at 5 and 7K, among the relative intensities of donor-acceptor and phonon emission peaks, a nonlinear relation has been seen and this fact has been proved by use of numerical data obtained in the experiment.

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